Use of municipal solid waste incineration bottom ash and crop by-product for producing lightweight aggregate

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Abstract. Due to the growing amount of residues in Europe, it is mandatory to provide a viable alternative for managing wastes contributing to the efficient use of resources. Besides, it is also essential to move towards a low carbon economy, priority EU by 2050. Among these, it is important to highlight the development of sustainable alternatives capable of incorporating different kind of wastes in their formulations.Municipal Solid Waste Incineration (MSWI) is estimated to increase in Europe, where the accessibility of landfill is restricted. Bottom ash (BA) is the most significant by-product from MSWI as it accounts for 85 - 95 % of the solid product resulting from combustion. BA is a mixture of calcium-rich compounds and others silicates enriched in iron and sodium. In addition, it is categorized as non-hazardous waste which can be revalorized as secondary material in construction or civil engineering fields, previous weathering stabilization during 2 - 3 months. Taking into account the relative proportion of each size fraction and the corresponding material characterization, the content of glass (primary and secondary) is estimated to be around 60 wt%. Furthermore, as a renewable resource and according to waste management European policies, residual agricultural biomass has attracted attention in preparation of advanced materials for various applications, due to their low cost, abundance, and environment friendliness. Among this residual biomass, rice husk is a by-product of rice milling industry which has high content of silica and has been widely used in buildings as natural thermal insulation material.Weathered BA (WBA) with a particle size less than 30 mm was milled under 100 μm, mixed with 2.0 - 5.0 mm rice husk, formed into ball-shaped pellets and sintered by different thermal treatments, which remove the organic matter content generating a large porosity. Physico-chemical analysis and mechanical behavior of the manufactured lightweight aggregates were tested. The obtained results provide a suitable physico-mechanical formulation using WBA as silica source, as well as a common crop by-product.

Keywords: Lightweight aggregate; Weathered Bottom Ash; Residual agricultural biomass; Rice husk.

1. Introduction

Incineration of municipal solid waste (MSW) in waste-to-energy plant is expected to increase all over the world because the increase in consumption and the use of landfills is limited [1]. Moreover, landfills are an option to avoid because of the production of methane, the soil and ground water contamination [2,3], the visual impact, and the necessity of recycling. Municipal solid waste incineration (MSWI) produces two types of ashes: BA, (bottom ash, around 17 Mt·year^{-1}) and Air Pollution Control fly ashes (APC) [4]. Although the incineration process considerably reduces the volume of MSW (by up to 90 %), 1 t of MSW produces approximately 0.3 t of BA, which is approximately 90 % of all residues generated during incineration process [5]. Besides, waste-to-energy plants allows to recover energy from municipal wastes, decreasing the use of fossil fuels and increasing the sustainability of the final management process.

BA is a heterogeneous material mainly composed of glass, synthetic and natural ceramics, magnetic and paramagnetic metals and a small percentage of unburned organic matter [6]. Glass is the major component (40 - 60 %), synthetic ceramics are around 26 %, and minerals compound are approximately 8 % [7]. The size distribution of BA is composed by 30 % of particles bigger than 6 mm, while 70 % by particles are bigger than 3 mm, and it is composed of both amorphous and crystalline phases, being Si the most abundant element mainly due to high lime-soda glass content. Si amount is followed by the presence of Ca, this from both synthetic and natural ceramic materials. BA is a silica-based material with the proper chemical and physical characteristics to be reused as secondary aggregate for civil engineering and construction, previous weathering stabilization (WBA) during 2-3 months [8,9]. Accordingly, nowadays, some countries in Europe are reusing their 50 % of the stockpiled MSWI BA, mainly for road construction or as a raw material for the construction material industry

[10–14]. Nevertheless, the reuse of BA can be restricted due to its high content in heavy metals, which are mainly concentrated in the particle size fraction lower than 4 mm [15,16].
Rice husk (RH) is an agricultural by-product comprising 40 % cellulose, 30 % lignin and 20 % silica [17] and, after combustion, its ash (RHA) is mainly composed of silica (> 90 %), most of which is amorphous depending on the temperature as well as the duration of the calcination process [13]. It is reported elsewhere [18] that RHA may be a residue with high economic potential, because it can be an alternative to alkali silicates (e.g. as raw material in the cement industri), while an agricultural by-product is valorized as energetic biomass.
Lightweight aggregates, which have very low densities (0.8 - 1.0 $g\cdot cm^{-3}$), are increasingly used to formulate lightweight concrete, lightweight geotechnical fill, insulation products, soil engineering, hydro-culture, drainage, roof gardens, and filters [19–21]. In addition, they have low thermal conductivity, as well as acoustic insulation and fire resistance [22]. Traditional lightweight aggregates are commonly produced using clay and, in some cases, adding oil to increase the porosity to provide to the material lower density [23]. However, because of its high glass content (i.e. low softening point material) and the possibility to encapsulate the heavy metals contained, which increasing its reuse, an attractive way to reuse WBA could be as a resource in the formulation of lightweight aggregates [24], while RH could be used to increase the porosity, providing in turn amorphous-Si to improve the cohesive forces between particles.
In this study, WBA is used as a matrix and RH as the biomass component to produce a final lightweight material. Although other studies use RH ashes or wood ashes [25], in this research the RH is incorporated directly in the sample, in order to produce high porosity during the combustion process. Accordingly, the aim of this research is to present the formulation, production, and testing of lightweight aggregate, formed into ball-shaped pellets, composed by WBA and RH. Characterization of the lightweight aggregate by means compression as mechanical property and shrinkage was also evaluated.

2. Experimental procedure

2.1. Materials

The solid precursor used for the present study derives from the < 30 mm fraction of municipal solid waste incineration weathered bottom ash (WBA). After removing metallic components by a magnet, WBA was milled and used in the lightweight formulations in the form of powder with grain size below 100 μm. The spherical grey samples (un-fired) were composed by WBA, RH, and deionized water, as can be seen in Figure 1.

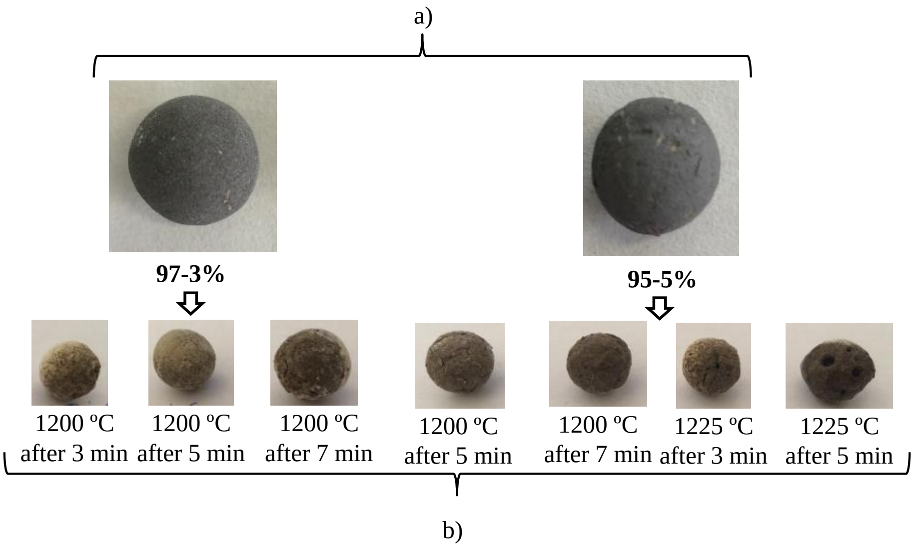


Figure 1. Image of the spherical lightweight formulations a) before, b) after sintering.

After formulating the handmade samples (with approximately 1 cm of diameter), they were dried at 105ºC during 24 h to constant weight. Subsequently, the un-fired specimens were put into a furnace at different temperatures (1175, 1200ºC, and 1225ºC), during different times (3, 5, 7, and 10 min). The different experimental sintering conditions are summarized in Table 1.

Table 1. Heating treatment and needed time for each formulation.

	WBA – RH (%, w/w)																			
Sintering conditions	97-3					95-5					90-10					85-15				
temperature (ºC)	1175	1200		1225		1175	1200		1225		1175	1200		1225		1175	1200		1225	
time (min)	10	5	7	3	5	10	5	7	3	5	10	5	7	3	5	10	5	7	3	5

The sintered specimens showed two different layers as showed in Figure 2: the outer one, with less porosity but more resistance and a darker color, and the inner one, with a high porosity and a clearer color. The samples also suffered a shrinkage that was measured.

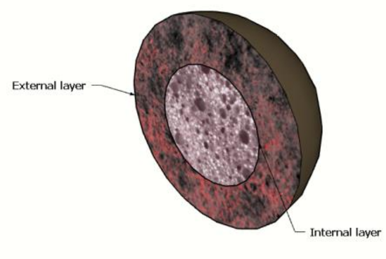


Figure 2. Lightweight aggregate with two different layers.

2.2. Methods

2.2.1. Physico-chemical characterization: X-Ray Fluorescence (XRF), X-Ray Diffraction (XRD), and Thermogravimetrical Analysis (TGA).

X-ray fluorescence semi-quantitative analysis was performed on the calcined WBA (550 ºC). A spectrophotometer Panalytical Philips PW 2400 sequential X-ray equipped with the software UniQuant® V5.0 was used.

With respect to the crystalline phases, these were determined by X-ray diffraction (XRD) for the WBA at 100 μm using a Bragg–Brentano Siemens D-500 powder diffractometer with CuKα radiation.

The thermal stability of the WBA and RH was evaluated with a Simultaneous SDTQ600 TA Instruments under N_2 and as well as in air atmosphere. The scanning rate was 10 K·min^{-1} in the temperature range between 25 and 1200 ºC.

2.2.2. Physico-mechanical characterization

The behavior of the test specimens was carried out using a universal testing machine from Zwick Roell. The specimens were inserted and the force (N) has been obtained. Then, the calculed compression strength was compared for each sample heating treatment. Denisty was measured considering the mass as well as the volume of each specimen, and shrinkage was evaluated following the Equation 1 (where V_i is the initial volume and V_f is the final volume). In addition, an observation was made by means of an optical microscope for the qualitative study of the internal porosity of the samples.

$$Shrinkage\ (\%) = \frac{V_i - V_f}{V_i} 100 \qquad \text{Equation 1}$$

3. Results and discussion

3.1. Physico-chemical characterization

The chemical composition of the major elements in the WBA by means of XRF is shown in Table 2. As can be observed, the XRF results agreed with the material characterization of WBA, which is mainly composed by lime-soda-glass, the most prevalent type of glass used in bottles.

Table 2. Chemical composition of MSWI weathered bottom ash (WBA).

Oxides	SiO_2	CaO	Cl	Fe_2O_3	Na_2O	Al_2O_3	MgO	K_2O	CuO	SO_3	ZnO
WBA (%)	49.38	14.69	n.d.	8.38	7.78	6.58	2.32	1.41	1.26	0.57	0.38

Figure 3 shows a diagram of WBA [27], where particle size distribution was determined by sieving the samples as indicated by the standard EN 933-2 [26].

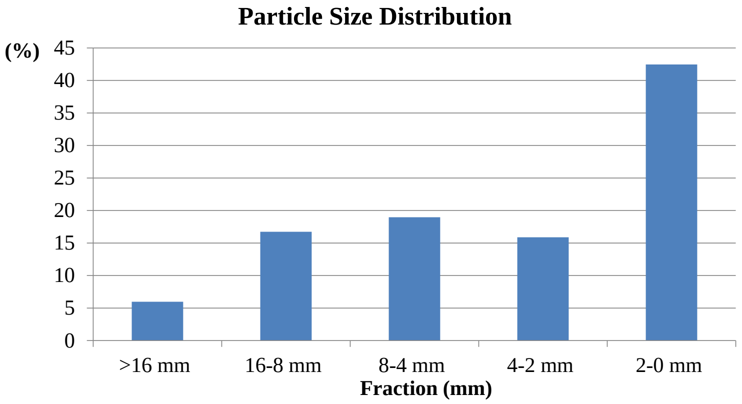
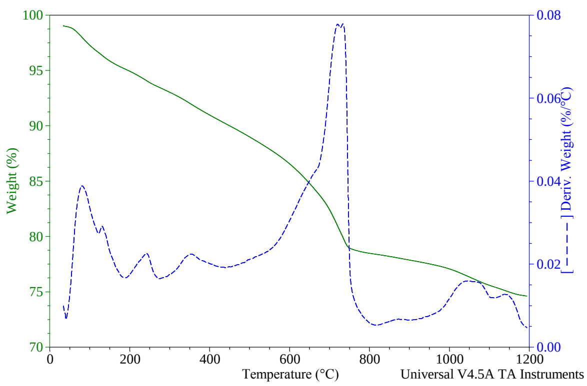


Figure 3. Particle size distribution for BA.

The XRD pattern performed to the WBA showed the presence of calcite $CaCO_3$, quartz SiO_2, hydroxylapatite $Ca_5(PO_4)_3(OH)$, dolomite $CaMg(CO_3)_2$, vaterite ($CaCO_3$), albite calcian low $(Na_{0.84}Ca_{0.16})Al_{1.16}Si_{2.84}O_8$, akermanite $Ca_2(Mg_{0.75}Al_{0.25})(Si_{1.75}Al_{0.25}O_7)$, orthoclase $KAlSi_3O_8$, yavapaiite $KFe(SO_4)_2$, and calcium sulfate $Ca(SO_4)$ as major crystalline phases. Hence, it is noticeable the important heterogeneity of the WBA sample as a function of particle size.

Taking into account the TG analysis, the thermal degradation for the WBA and RH can be observed in Figure 4. For the WBA sample, the heating ramp was 10 ºC·min^{-1} between 30 and 1200 ºC using 50 mL·min^{-1} of air atmosphere. On the other hand, the rice husk was analyzed at 5 ºC·min^{-1} between 30 and 1200 ºC and with 50 mL·min^{-1} of air atmosphere. As it can be observed, the WBA sample has four main steps with a total decomposition of 25 %, which denote the presence of organic matter, carbonates, and other hydroxides and hydrated salts, decomposing with temperature. For the rice husk analysis (Figure 4b), it can be observed that around 800 ºC that all the sample is decomposed, having three different steps. The first 5 % mass decrease occurred between 20 and 105 ºC due to the evaporation of water moisture [28,29]. The second and third mass loss was observed in the 200 - 450 ºC range is due to the rice husk burning, were mainly two large decompositon reactions between 200 and 500 ºC as it is clearly shown by two peaks in Derivative weight signal (discontinous blue line in Figure 4), these decompositions correspond to the burning of rice husk.



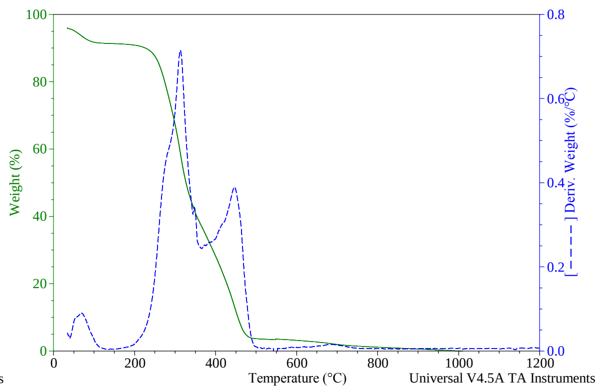


Figure 4. TGA result from a) WBA (fraction >30 mm urban waste and milled until 100 μm) and b) RH.

3.2. Physico-mechanical characterization

Once the sintered aggregates were obtained, the mechanical characterization was performed to each one. The compressive strength values of the lightweight aggregates between the different proportions WBA-RH were determined. The results are shown in Figure 5, where the highest value for the compressive strength relays at 1200ºC during 7 minutes for the 95-5% lightweight aggregate. It is important to highlight that, for a similar mechanical behavior, it is expected that a greater amount of rice husk give rise to a greater porosity and, therefore, a better lightweight aggregate. Hence, as the result for the 95-5 % at 1200 ºC during 7 minutes is relatively close to the value for the 85-15% at 1200ºC during 7 minutes, it can be considered that, the best lightweight aggregate from the total formulated samples is 85-15% (WBA-RH) at 1220 ºC during 7 minutes regarding the compression as the mechanical property evaluated. Besides, in Figure 6 is showed the density and shrinkage results of all the samples under study. As it was expected, it can be observed that the lowest values for the density parameter are those regarding the 85-15 % formulation. Considering the shrinkage of the samples, the results have several fluctuations; this fact can be attributed to the handmade preparation of the samples. Nevertheless, it is very interesting to corroborate that the shrinkage for the sample 95-5 % at 1200 ºC during 7 min as a heatig treatment has approximately the same value as the 85-15 at 1200 ºC during 7 minutes. This results are in agreement with those explained for the compression assay.

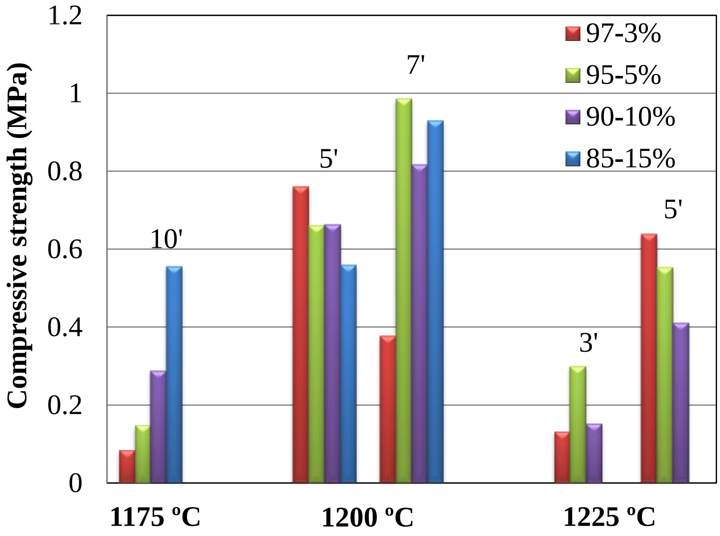


Figure 5. Compressive strength results.

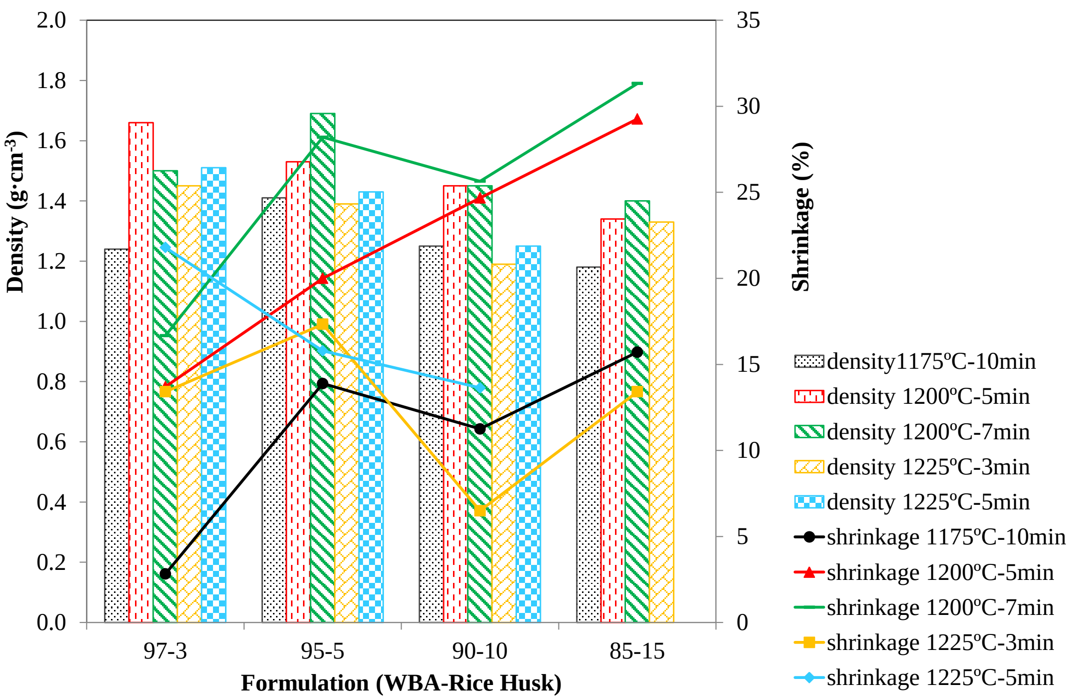
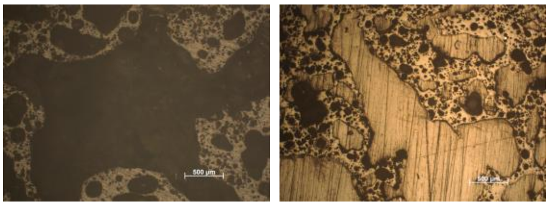
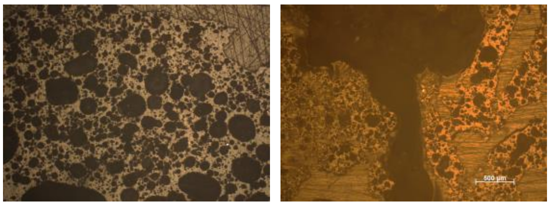


Figure 6. Density and shrinkage results for all the samples under study.

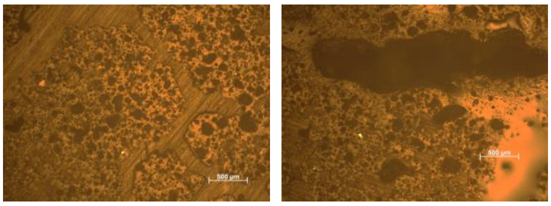
In Figure 7 is shown the micrographies from the optical microscope observation. As it can be seen, as higher content of RH higher porosity.



85-5% 1200ºC 7min 85-5% 1175ºC 10min



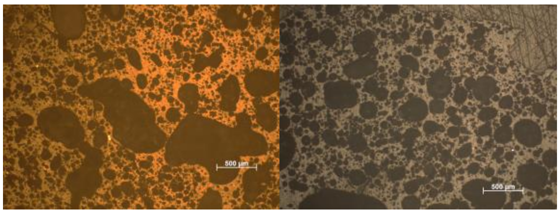
95-5% 1200ºC 7min 95-5% 1175ºC 10min



97-3% 1200ºC 7min 97-3% 1175ºC 10min

Figure 7. comparison of micrograpahies from different composition, temperatures and time of sinterization.

Besides, in the micrographies of Figure 8, it can be observed that (from the same temperature) as higher curing time, the porous is lower.



95-5% 1200ºC 5 min 95-5% 1200ºC 7 min

Figure 8. Porous comparison between two samples at the same temperature, but different sintered time.

4. Conclusions

It has been formulated a new lightweight material composed by municipal solid waste incineration weathered bottom ash (WBA) and rice husk (RH) as biomass. Different proportions and heating treatments, and different curing times were evaluated. Compressive streght of the lightweight aggregates was determined, where it was observed that the highest value for the compressive strength relays in the 85-15 % at 1220ºC during 7 minutes. In addition, by means optical microscopy, it was evaluated the porosity, concluding that as higher content of RH higher porosity. Then, it can be deduced that the formulation 85-15 % WBA-RH is the best formulation, although a leaching potential analysis should be performed to confirm the application of this lightweight material in secondary aggregates. Then, further analysis will be performed to corroborate that the formulated material has the chemical-physical-mechanical and environmental requirements.

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